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(54) THERMOPLASTIC ELASTOMER COMPOSITION

(57)Abstract:

**PROBLEM TO BE SOLVED:** To obtain a thermoplastic elastomer composition having excellent balance of mechanical strength and elastic recovery and excellent in flexibility and fluidity, and useful as interior and exterior members of an automotive by dynamically heat-treating a specific composition in the presence of a crosslinking agent for rubber.

**SOLUTION:** This thermoplastic elastomer composition is obtained by dynamically heat-treating a composition comprising (A) 10-90wt.% of olefinic copolymer rubber such as ethylene-propylene copolymer rubber, (B) 1-80wt.% of a hydrogenated diene-based polymer, (C) 180wt.% of a crystalline  $\alpha$ -olefinic polymer containing a  $\geq 3C$   $\alpha$ olefin as a main component, (D) 1-80wt.% of an ethylenic polymer (a total of the components A to D is 100wt.%) and (E) 0-200pts.-wt. of a mineral oil-based softening agent such as a naphthene oil to 100 pts.wt. of a total amount of the components A to D in the presence of a crosslinking agent for rubber such as dicumyl peroxide.

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CLAIMS

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[Claim(s)]

[Claim 1] (b) 10 - 90 % of the weight of olefin system copolymerization rubber, 1 - 80 % of the weight of (b) hydrogenation diene system polymers, 1 - 80 % of the weight of crystalline alpha olefin system polymers which use a with a carbon numbers of three or more alpha olefin as a principal component, (Ha) (\*\*) — 1 - 80 % of the weight of ethylene system polymers — [ — however, (\*\*) — + (\*\*) — +(Ha)+ (d) =100 % of the weight] — And the thermoplastic-elastomer constituent which comes dynamically to heat-treat the constituent which blended the 0 - 200 weight section for the (e) straight-mineral-oil system softener to the total quantity 100 weight section of an above-mentioned (b) - (d) component under existence of a rubber frame-common-equipment pons agent.

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**DETAILED DESCRIPTION**

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**[Detailed Description of the Invention]**

**[0001]**

**[Field of the Invention]** This invention relates to the thermoplastic-elastomer constituent which blends a hydrogenation diene system polymer and an ethylene system polymer at the crystalline alpha olefin system copolymer which uses a with olefin system copolymerization rubber and a carbon numbers of three or more alpha olefin as a principal component at a detail, and is obtained by heat-treating dynamically under existence of a rubber frame-common-equipment pons agent and which has high intensity and was flexible and was rich in the fluidity further about a thermoplastic-elastomer constituent.

**[0002]**

**[Description of the Prior Art]** Elastic vinyl chloride system resin (PVC) has the good balance of a mechanical strength and flexibility, although it is the ingredient excellent in cost performance, the use is restricted from the viewpoint of earth environment, and the alternative to the thermoplastic elastomer of an olefin system etc. is progressing [ the part ]. Conventionally, the thermoplastic-elastomer-olefin constituent is obtained by stiffening a rubber part dynamically by the known approach. It is possible to obtain the thermoplastic-elastomer constituent excellent in elastic recovery nature, without spoiling the cold resistance which EPM and/or EPDM have, weatherability, and chemical resistance by this approach. However, its compatibility of olefin system resin and olefin system rubber is inadequate, and although the above thermoplastic-elastomer constituents over which the bridge was constructed dynamically have constructed the bridge over a rubber part, they are difficult to obtain sufficient mechanical strength.

**[0003]**

**[Problem(s) to be Solved by the Invention]** This invention was made against the background of the technical problem of the above-mentioned conventional technique, and aims at offering the thermoplastic-elastomer constituent which was able to balance the mechanical strength to elastic recovery nature.

**[0004]**

**[Means for Solving the Problem]** This invention 10 - 90 % of the weight of (b) olefin system copolymerization rubber, 1 - 80 % of the weight of (b) hydrogenation diene system polymers, 1 - 80 % of the weight of crystalline alpha olefin system polymers which use a with a carbon numbers of three or more alpha olefin as a principal component, (Ha) (\*\*) -- 1 - 80 % of the weight of ethylene system polymers -- [ -- however, (\*\*) -- + (\*\*) -- + (Ha) + (d) = 100 % of the weight ] -- And the thermoplastic-elastomer constituent which comes dynamically to heat-treat the constituent which blended the 0 - 200 weight section for the (e) straight-mineral-oil system softener to the total quantity 100 weight section of an above-mentioned (b) - (d) component under existence of a rubber frame-common-equipment pons agent is offered.

**[0005]**

**[Embodiment of the Invention]** As (b) olefin system copolymerization rubber of this invention, the elastic copolymer of the amorphism which uses a with a carbon numbers [ like ethylene propylene copolymerization rubber, an ethylene propylene and nonconjugated diene ternary polymerization rubber, ethylene and 1-butene copolymerization rubber, and ethylene, 1-butene and nonconjugated diene ternary polymerization rubber / the ethylene and three or more carbon numbers ] alpha olefin as a principal component, for example is mentioned.

**[0006]** As the above-mentioned nonconjugated diene, for example 1, 4-pentadiene, 1, 4-hexadiene, 1, 5-hexadiene, 1, 7-OKUTA diene, 1, 9-deca diene, 3, the 6-dimethyl -1, 7-OKUTA diene, 4, the 5-dimethyl -1, 7-OKUTA diene, The 5-methyl -1, 8-nonadiene, a dicyclopentadiene, 5-ethylidene norbornene, 5-vinyl-2-norbornene, 2, and 5-norbornadiene etc. can be mentioned, and 1, 4-hexadiene, a dicyclopentadiene, and 5-ethylidene-2-norbornene are especially desirable.

**[0007]** In these (b) olefin system copolymerization rubber, it is desirable that copolymerization of the with a carbon numbers of three or more alpha olefin is carried out at 10% of the weight or more of a rate. Moreover, as nonconjugated diene, ethylidene norbornene, dicyclopentadiene, 1, and 4-hexadiene is desirable, and it is desirable an amount which becomes 40 or less by iodine number display, and to exist.

(b) In olefin system copolymerization rubber, the content of an ethylene component increases more than 90 % of the weight, and the content of an alpha olefin component runs short [ the flexibility of this copolymerization rubber ] and is not desirable at less than 10 % of the weight. moreover, (\*\*) -- as for the Mooney viscosity (ML 1+4, 100 degrees C) of olefin system copolymerization rubber, it is desirable 10-500, and that it is 30-400 preferably. If this Mooney viscosity is smaller than 10, reinforcement will become low, and on the other hand, if larger than 500, a maldistribution with (\*\*) and (Ha) a (d) component generates and is not desirable.

**[0008]** (b) Although especially the polymerization method of olefin system copolymerization rubber is not restricted, a polymerization is carried out under existence of a catalyst with well-known vanadium system catalyst, titanium system catalyst, metallocene system catalyst, etc. Moreover, a part of hydrogen atom of (b) olefin system copolymerization rubber receives the

halogenation copolymerization rubber permuted by halogen atoms, such as a chlorine atom and a bromine atom, or this olefin system copolymerization rubber, and halogenation copolymerization rubber. a vinyl chloride, vinyl acetate, an acrylic acid (meta), or its derivative — [ — for example ], such as methyl (meta) acrylate, glycidyl (meta) acrylate, and acrylamide (meta), Partial saturation monomers, such as a maleic acid or its derivatives (for example, a maleic anhydride, maleimide, maleic-acid dimethyl, etc.), and conjugated dienes (for example, a butadiene, an isoprene, a chloroprene, etc.), can also use the graft copolymer which carried out graft polymerization. Using it by the one-sort independent can also be mixed and used for these (b) olefin system copolymerization rubber for two or more sorts.

[0009] Next, a (b) hydrogenation diene system polymer hydrogenates the conjugated diene system polymer which makes conjugated diene a subject. For example, the homopolymer of conjugated diene, the random copolymer of conjugated diene and an aromatic series vinyl compound, The block copolymer which consists of a polymer block of an aromatic series vinyl compound, and a polymer block of conjugated diene, The block copolymer which consists of a polymer block of an aromatic series vinyl compound, and a copolymer block of conjugated diene / aromatic series vinyl compound, The hydrogenation object of conjugated diene system polymers, such as copolymer blocks which consist of a polymer block of conjugated diene and a copolymer block of conjugated diene / aromatic series vinyl compound, or these functional-group denaturation objects, is mentioned.

[0010] these various hydrogenation objects — each — (\*\*\*) of this invention — although suitably used as a component, especially the following (low 1), (low 2), or (low 3) hydrogenation diene system polymers that are shown are especially used suitably as a (b) component. (\*\*\*) of the following this inventions — the hydrogenation diene system polymer (low 1) shown below, (low 2), and (low 3) are made into an example, and explanation of a hydrogenation diene system polymer explains them.

[0011] (\*\*-1) A component (low 1) component is a hydrogenation diene system polymer obtained by hydrogenating the polymer which makes a subject the random copolymerization parts of conjugated diene and an aromatic series vinyl compound.

[0012] As conjugated diene used for a component (low 1) here 1,3-butadiene, an isoprene, 2,3-dimethyl-1,3-butadiene, Although 1,3-pentadiene, 2-methyl-1,3-pentadiene, 1, 3-hexadiene, 4, the 5-diethyl -1, 3-OKUTA diene, 3-butyl -1, 3-OKUTA diene, a chloroprene, etc. are mentioned In order to obtain the hydrogenation diene system polymer which could use industrially and was excellent in physical properties, 1,3-butadiene, an isoprene, and 1,3-pentadiene are desirable, and 1,3-butadiene and especially an isoprene are desirable.

[0013] moreover, as an aromatic series vinyl compound, styrene, alpha-methyl-styrene, p-methyl styrene, t-butyl styrene, divinylbenzene, N, and N-dimethyl-p-aminoethyl styrene, N, and N-diethyl-p-aminoethyl styrene, vinylpyridine, etc. are mentioned, it is styrene and alpha methyl styrene more preferably, and these are one-sort independent — it is — two or more sorts can be used, using together.

[0014] In the polymer before hydrogenation, the rate in the inside of the polymer before hydrogenation of a random copolymerization part is 60 % of the weight or more still more preferably 50% of the weight or more preferably, and flexibility falls at less than 50 % of the weight. Moreover, it is unsaturated bond [ 1 and 2-association, and 3 and 4 to a side chain [ on the above-mentioned random copolymerization part and as opposed to all the conjugated diene units in a random copolymerization part ]. - It joins together (15% or more, it is 20% or more still more preferably, and the rate of the conjugated diene which has] called "vinyl association" below does not have flexibility enough for less than 15% of case, and it is not preferably desirable to it.). Although the rate of the conjugated diene / aromatic series vinyl compound which constitutes the polymer before hydrogenation is not limited especially in this invention, they are 93 / 7 - 50/50 % of the weight still more preferably preferably five to 40/60 95-% of the weight.

[0015] Although the component (low 1) of this invention is obtained by hydrogenating the polymer which makes a random copolymerization part a subject as mentioned above, the following polymer blocks may be included in the chain of the polymer before hydrogenation. It consists of an aromatic series vinyl compound polymer, 1, the polybutadiene polymer that makes 4-association a subject and an aromatic series vinyl compound, and conjugated diene as a polymer block which may be included in the polymer before hydrogenation, and the taper-like polymer which an aromatic series vinyl compound increases gradually is mentioned. Since handling nature will improve when the blocking nature of an ingredient falls although the description on the physical properties of a component (low 1) is spoiled a little if these polymer blocks exist, it may be industrially useful. Although especially the rate in all the chains of the above-mentioned polymer block in the polymer before hydrogenation is not limited, its 50 or less % of the weight is desirable, and its 40 or less % of the weight is still more desirable. The fall of flexibility becomes large when exceeding 50 % of the weight.

[0016] The above-mentioned polymer before hydrogenation may be a polymer to which the polymer chain extended or branched through coupling agent residue by use of a coupling agent. As a coupling agent in this case, adipic-acid diethyl, a divinylbenzene, methyl dichlorosilane, tetrachlorosilane, buthyl trichloro silicon, tetra-chloro tin, BUCHIRUTORI chloro tin, dimethyl dichloro silicon, tetra-chloro germanium, 1,2-dibromoethane, 1, 4-chloro methylbenzene, screw (trichlorosilyl) ethane, the epoxidation linseed oil, tolylene diisocyanate, 1 and 2, 4-benzene tri-isocyanate, etc. are mentioned, for example.

[0017] in the hydrogenation (low 1) diene system polymer of this invention, the rate of hydrogenation of the double bond of the conjugated diene part in a chain is still more desirable preferably 80% or more — it is 90% or more especially preferably 85% or more. At less than 80%, weatherability becomes inadequate. Moreover, number average molecular weight will be 50,000-600,000 preferably [ it is desirable and ] to 50,000-700,000, and a pan, and less than by 50,000, the hydrogenation (low 1) diene system polymer of this invention has an inadequate mechanical strength, on the other hand, if 700,000 is exceeded, it will run short of a fluidity and will become what has inadequate fabrication nature. (\*\*-1) A component can be obtained by the approach currently indicated by JP,3-72512,A.

[0018] (\*\*-2) A component (low 2) component is the hydrogenation diene system polymer which hydrogenated the block copolymer which uses each block of the following (A), (B), and (C) as a principal component.

[0019] The aromatic series vinyl compound used here in order to obtain a component (low 2), and conjugated diene are the same as that of what is used in order to obtain the above-mentioned (low 1) component. (\*\*-2) The desirable (A) block which

constitutes a hydrogenation diene system polymer is a polymer block which makes an aromatic series vinyl compound a subject, and is a polymer block with which hydrogenation of the 80% or more of the conjugated diene part of the homopolymer of an aromatic series vinyl compound or a copolymer with the conjugated diene which has an aromatic series vinyl compound 90% of the weight or more during the (A) block was carried out to the detail. (A) Reinforcement and weatherability fall [ the aromatic series vinyl compound under block ] at less than 90 % of the weight. (\*\*-2) The desirable content of the (A) block in a component is 10 - 55 % of the weight still more preferably five to 60% of the weight. At less than 5 % of the weight, thermal resistance and a mechanical strength are inferior, and on the other hand, when it exceeds 60 % of the weight, workability and flexibility are inferior. moreover, the desirable number average molecular weight of the (A) block — 02000— it is 420,000.

[0020] Moreover, 30 to 90% of the weight, it is 35 - 80 % of the weight still more preferably, and at less than 30 % of the weight, if flexibility, on the other hand, exceeds 90 % of the weight by falling, as for the desirable content of the (B) block which constitutes a hydrogenation (low 2) diene system polymer, workability and a mechanical strength will fall. (B) The vinyl joint content of the conjugated diene part before the hydrogenation included in a block is 30 - 90% still more preferably 25 to 95% preferably. (B) If hydrogenation of the vinyl joint content is carried out at less than 25% when conjugated diene is a butadiene among the conjugated diene blocks before the hydrogenation which is blocked, a polyethylene chain will generate, a rubber-property will be lost, if it exceeds 95% on the other hand and hydrogenation will be carried out, glass transition temperature will become high, a rubber-property is lost, and it is not desirable. (B) the desirable number average molecular weight of a block — desirable — 15,000- 630,000 — further — desirable — 35,000- it is 420,000. Moreover, the rate of hydrogenation of the double bond of the conjugated diene part of the (B) block is 80% or more.

[0021] Furthermore, the vinyl joint content of the (C) block which constitutes the polymer before hydrogenation for obtaining a component (low 2) is less than 20% of polybutadiene polymer block preferably less than 25%. If hydrogenation of the vinyl joint content is carried out at 25% or more, a resin-property will be lost and the property of the thermoplastic elastomer as a block copolymer will be lost. The content of the (C) block in the above-mentioned polymer before hydrogenation is 5 - 50 % of the weight preferably five to 60% of the weight. (C) If the mechanical property of a component (low 2) is inferior in the content of a block and it, on the other hand, exceeds 60 % of the weight at less than 5 % of the weight, a rubber-property is lost and it is not desirable. (C) the desirable number average molecular weight of a block — 02500- it is 420,000 and the rate of hydrogenation of the double bond of the butadiene part of a polybutadiene block is 80% or more.

[0022] The polymer before hydrogenation which constitutes (low 2) may be a block copolymer with which a polymer chain which is expressed with the following general formula extended or branched by adding a coupling agent.

$[(A)-(B)-(C))_n-X$  or  $[(A)-(B)-(C))_n-[X](A)-(B)-]$

(A), (B), and (C) are the same as the above among [type. n shows the integer of 2-4 and X shows coupling agent residue. ]

As a coupling agent in this case, it is the same as that of what is used of the above-mentioned (low 1) component.

[0023] By carrying out hydrogenation of the polymer before hydrogenation which constitutes the above hydrogenation (low 2) diene system polymer, when the double bond of the conjugated diene part of this polymer is saturated, the component which is a hydrogenation diene system polymer (low 2) is obtained. Here, the double bond of conjugated diene needs to carry out hydrogenation of the 80% or more, and is 95 - 100% still more preferably 90% or more preferably. The rate of hydrogenation of the double bond of a conjugated diene part is inferior in the thermal stability of a thermoplastic-elastomer constituent, and endurance at less than 80%. Moreover, the number average molecular weight of a component (low 2) is 50,000-700,000, and is 100,000-600,000 preferably. By less than 50,000, thermal resistance, reinforcement, a fluidity, and workability fall, and on the other hand, when 700,000 is exceeded, a fluidity, workability, and flexibility are inferior. (\*\*-2) A component can be obtained by the approach currently indicated by JP,2-133406,A.

[0024] (\*\*-3) A component (low 3) hydrogenation diene system polymer (D) The polybutadiene polymer block whose vinyl joint content is 25% or less, (E) They are a conjugated diene polymer block or a copolymer block with an aromatic series vinyl compound and conjugated diene. the polymer block whose vinyl joint content of a conjugated diene part is 25 - 95% — (D)-(E)-(D) or (D)- it is obtained by hydrogenating the double bond part of the block copolymer of the shape of a straight chain arranged as shown in (E), and the letter of branching 80% or more.

[0025] The aromatic series vinyl compound used here in order to obtain a component (low 3), and conjugated diene are the same as that of what is used for a component (low 1). (\*\*-3) The (D) block in a component turns into a crystalline polymer block which shows structure similar to usual low density polyethylene (LDPE) by hydrogenation. (D) Although the vinyl joint content under block is 25% or less, it is usually 15% or less still more preferably 20% or less preferably. (D) When the vinyl joint content under block exceeds 25%, hardening of the crystalline melting point after hydrogenation is remarkable, and a mechanical property is inferior.

[0026] Moreover, the (E) block is a conjugated diene polymer block or an aromatic series vinyl compound-conjugated diene copolymer block, and turns into a rubber-like block [ ethylene-butene-1 copolymer ] or a polymer block which shows structure similar to an aromatic series vinyl compound-ethylene-butene-1 copolymer by hydrogenation.

[0027] In addition, the monomer which constitutes the (E) block is 25 or less % of the weight still more preferably 30 or less % of the weight preferably 35 or less % of the weight, the glass transition temperature of the (E) block rises and the amount of the aromatic series vinyl compound used for the (E) block is inferior in a low-temperature property and flexibility, when it exceeds 35 % of the weight. Moreover, 25 to 95%, 25 to 75%, when conjugated diene is a butadiene, it shows the crystal structure originating in a polyethylene chain and polybutene -1 chain, and serves as resin-like description, and it is preferably inferior [ content / the vinyl joint content of the conjugated diene part of the (E) block is 25 - 55%, and / with hydrogenation ] in flexibility still more preferably, when it exceeds less than 25% or 95%, respectively.

[0028] moreover, the polymer before hydrogenation which constitutes a hydrogenation (low 3) diene system polymer — setting — the rate of the (D) block and the (E) block — usually — it is [ 5 - 90 % of the weight of (D) blocks ] 90 - 20 % of the weight [however, (D)+(E) =100 % of the weight] preferably 80% of the weight 95 - 10 % of the weight of 10 - (E) blocks. (D) Since crystalline polymer blocks run short and the mechanical property of a component (low 3) is inferior when the (E) block exceeds

[ a block ] 95 % of the weight less than 5 % of the weight, it is not desirable. Moreover, the (D) block exceeds 90 % of the weight, and when the (E) block is less than 10 % of the weight, the degree of hardness of a component (low 3) rises, and it is not desirable. in addition, the desirable number average molecular weight of the (D) block — 02500— 630,000 — it is 10,000—480,000 still more preferably. Moreover, the desirable number average molecular weight of the (E) block is 20,000—540,000 preferably to 05000 — 665,000, and a pan.

[0029] In addition, the polymer before hydrogenation which constitutes a hydrogenation (low 3) diene system polymer may be a block copolymer with which a polymer chain which is expressed with the following general formula extended or branched by adding a coupling agent.

$[(D)-(E)]_n-X$  — or —  $[-(D)-(E)-(D)-]_n-X$  — [ — a formula — inside —  $-(D)-(E)-$  — the above — being the same . n shows the integer of 2-4 and X shows coupling agent residue. ]

As a coupling agent in this case, it is the same as that of what is used for the above-mentioned (low 1) component.

[0030] By carrying out hydrogenation of the above polymer before hydrogenation, the component which is a hydrogenation diene system polymer (low 3) is obtained by carrying out hydrogenation of the double bond of the conjugated diene part of this polymer. Here, the double bond of conjugated diene needs to carry out hydrogenation of the 80% or more, and is 95 — 100% still more preferably 90% or more preferably. The rate of hydrogenation of the double bond of a conjugated diene part is inferior in the thermal stability of thermoplastic elastomer, and endurance at less than 80%. moreover, the number average molecular weight of a component (low 3) — 50,000—700,000 — it is 100,000—600,000 preferably. By less than 50,000, thermal resistance, reinforcement, a fluidity, and workability fall, and on the other hand, when 700,000 is exceeded, a fluidity, workability, and flexibility are inferior. (\*\*-3) A component can be obtained by the approach currently indicated by JP,3-1289576.A.

[0031] The denaturation hydrogenation diene system polymer which denaturalized by the functional group is sufficient as the (b) hydrogenation diene system polymer used for this invention. This denaturation hydrogenation diene system polymer makes a hydrogenation diene system polymer come to contain at least one sort of functional groups chosen from the group of a carboxyl group, an acid-anhydride radical, hydroxyl, an epoxy group, a halogen atom, the amino group, an isocyanate radical, a sulfonyl group, and a sulfonate radical. As an approach of making this functional group containing, the conjugated diene or the aromatic series vinyl compound which has \*\* functional group is used. Copolymerize, where the functional group of a monomer is protected, and a block copolymer is obtained. The approach of making it add during a polymerization by the technique of performing deprotection after a polymerization conclusion, the method of making the radical polymerization nature monomer which has \*\* functional group add to a hydrogenation diene system polymer by the known graft-ized reaction, \*\* Using the monomer containing a functional group, under existence of organic peroxide or an azo compound or nonexistence, a kneader, a mixer, an extruder, etc. are used, a hydrogenation diene system polymer is kneaded, and the approach to which a functional group is made to add is mentioned.

[0032] Although a functional group can be made to contain efficiently even if it uses which these approaches, industrially, the approach of the above-mentioned \*\* — \*\* is simple, and effective. the molecule with which the content of the functional group in this denaturation hydrogenation diene system polymer usually constitutes a hydrogenation diene system polymer — receiving — 0.01-10-mol % — desirable — 0.1-8-mol % — it is 0.15-5-mol % still more preferably. As a desirable example of the monomer which adds a functional group to a hydrogenation diene system polymer, an acrylic acid, a methacrylic acid, an itaconic acid, a maleic acid, a maleic anhydride, metaglycidyl acrylate, glycidyl methacrylate, allyl glycidyl ether, hydroxy ethylene glycol, hydroxypropyl methacrylate, hydroxyethyl acrylate, hydroxypropyl acrylate, dimethylaminoethyl methacrylate, etc. are mentioned.

[0033] Next, as a crystalline alpha olefin system polymer which uses as a principal component a with a carbon numbers of three or more used for the constituent of this invention (Ha) alpha olefin, polypropylene, poly1 butene, poly4 methyl 1 pentene, a Pori 1-hexene, a propylene-ethylene copolymer, a propylene-1-butene copolymer, etc. are mentioned. Polypropylene, random, or block ethylene propylene rubber is [ among these ] desirable.

[0034] (c) A component is crystallinity and n-Deccan insoluble matter is specifically 70% of the weight or more of a thing still more preferably 60% of the weight or more preferably 50% of the weight or more. If n-Deccan insoluble matter uses less than 50% of the weight of a thing, the mechanical strength of the thermoplastic-elastomer constituent obtained and fabrication nature may be spoiled.

[0035] Next, as a (d) ethylene system polymer used for this invention, polyethylene or ethylene, and a carbon number may be the copolymers of a copolymer with alpha olefins, such as the propylene of 3-10, butene-1, 4-methyl, a pentene -1, a hexene -1, and octene, or ethylene, a vinyl acetate, an acrylic acid, etc., etc. (d) Although especially the polymerization method of a component is not restricted, the polymerization of it is carried out under existence of a catalyst with well-known vanadium system catalyst, titanium system catalyst, metallocene system catalyst, etc. In addition, as for a (d) component, it is desirable that an ethylene content is 90 % of the weight or more. The mechanical strengths of the constituent with which an ethylene content is obtained at less than 90 % of the weight run short.

[0036] The loadings of the above-mentioned (\*\*) used for the constituent of this invention — (Ha) a component (b) olefin system copolymerization rubber preferably ten to 90% of the weight 30 — 80 % of the weight. A (b) hydrogenation diene system polymer 50 to 75% of the weight still more preferably 1 — 80 % of the weight. Still more preferably 20 to 60% of the weight preferably 30 — 60 % of the weight. The crystalline alpha olefin system polymer which uses a with a carbon numbers of three or more alpha olefin as a principal component 1 — 80 % of the weight, (Ha) desirable — 3 — 50 % of the weight — further — desirable — 5 — 40 % of the weight, and (\*\*) — an ethylene system polymer — 1 — 80 % of the weight — desirable — 3 — 50 % of the weight — further — desirable — 5 — 40 % of the weight — [ — however, (\*\*) — + (\*\*) — it is +(Ha)+ (d) =100 % of the weight].

[0037] Here, the rubber elasticity and flexibility of a constituent which are acquired as the loadings of (b) olefin system copolymerization rubber are less than 10 % of the weight come to get worse. On the other hand, if a (b) component exceeds 90 % of the weight, the fabrication nature of the constituent obtained and a mechanical strength will get worse. Moreover, the elastic recovery nature and the mechanical strength of a constituent from which a (b) component is obtained at less than 1 % of the

weight are inferior, and on the other hand, when it exceeds 80 % of the weight, the thermal resistance and the fluidity of a constituent which are acquired are inferior. Furthermore, the thermal resistance of the constituent obtained as a component (Ha) is less than 5 % of the weight gets worse, and on the other hand, if it exceeds 80 % of the weight, the flexibility of the constituent obtained will get worse. Furthermore, the elastic recovery nature and the mechanical strength of a constituent from which a (d) component is obtained at less than 1 % of the weight are inferior, and on the other hand, if it exceeds 80 % of the weight, the flexibility of the constituent obtained runs short.

[0038] Next, as a straight-mineral-oil system softener of a (e) component used for this invention, a naphthene oil and paraffin series straight mineral oil can be used. By such oil exhibition, workability and flexibility improve further. in this case, \*\*\*\*\* — the total quantity 100 weight section of an above-mentioned (b) - (d) component — receiving — the 0 - 200 weight section — desirable — the 0 - 100 weight section — it is 0 - 50 weight section still more preferably. (e) When \*\*\*\*\* of a straight-mineral-oil system softener exceeds the 200 weight sections, while the mechanical strength of the constituent obtained is inferior, since the component of straight mineral oil deposits on a constituent front face, it is not desirable.

[0039] Under existence of a rubber frame-common-equipment pons agent, the thermoplastic-elastomer constituent of this invention is heat-treated dynamically, and the constituent which uses as a principal component an above-mentioned (b) - (d) component and the (e) component further blended with these components if needed is obtained.

[0040] Although especially the rubber frame-common-equipment pons agent used for this invention is not limited, it is desirable to usually use the cross linking agent used for vulcanization of olefin system copolymerization rubber. A thing desirable in it is organic peroxide or a phenol system resin cross linking agent.

[0041] As organic peroxide, for example Among these, dicumyl peroxide, Di-*t*-butyl peroxide, 2, the 5-dimethyl -2, 5-di-*tert*-butyl peroxide hexane, 2, 5-dimethyl-di-*tert*-butyl peroxide hexyne - 3, 1, 3-screw (*t*-butyl PAOKI seesaw propyl) benzene, 1 and 1-screw (*tert*-butyl peroxide) - 3, 3, a 5-trimethyl cyclohexane, *n*-butyl -4, 4-screw (*tert*-butyl peroxide) valerate, benzoyl peroxide, *p*-KURORU benzoyl peroxide, 2, 4-dichloro benzoyl peroxide, *T*-butyl peroxybenzoate, *t*-butylperoxyisopropylcarbonate, diacetyl peroxide, lauroyl peroxide, *t*-butyl peroxide, etc. are mentioned.

[0042] Also in these organic peroxide, after distributing rubber and a resinous principle more The organic peroxide which carries out decomposition initiation is desirable, for example, it is 2 and 5-dimethyl. - 2, 5-di-*tert*-butyl peroxide-hexyne - 3, 2, the 5-dimethyl -2, 5-di-*tert*-butyl peroxide hexane, 1 and 3-screw (*t*-butyl PAOKI seesaw propyl) benzene is mentioned, and 2 with especially high decomposition temperature, the 5-dimethyl -2, and 5-di-*tert*-butyl peroxide-hexyne -3 are the most desirable.

[0043] If a suitable bridge formation assistant is made to exist with organic peroxide, homogeneity and crosslinking reaction [ \*\*\*\* ] are expectable. As a bridge formation assistant, sulfur, *p*-quinonedioxime, *p*, and *p*'-dibenzoyl quinonedioxime, Ethylene glycol dimethacrylate, diethylene-glycol dimethacrylate, Triethylene glycol dimethacrylate, tetra-ethylene dimethacrylate, Polyethylene glycol dimethacrylate, trimethylolpropanetrimethacrylate, Diaryl phthalate, diaryl phthalate, tetra-aryloxy ethane, Thoria reel SHIANU rate, diaryl phthalate, tetra-aryloxy ethane, thoria reel SHIANU rate, N, and N-*m*-phenylene bismaleimide, a maleic anhydride, a divinylbenzene, JI (meta) acrylic-acid zinc, etc. are used. Preferably, it is desirable to use N and N-*m*-phenylene bismaleimide, *p*, and *p*'-dibenzoyl quinonedioxime and a divinylbenzene. Moreover, it can also use as a cross linking agent by the N and N-*m*-phenylene bismaleimide independent.

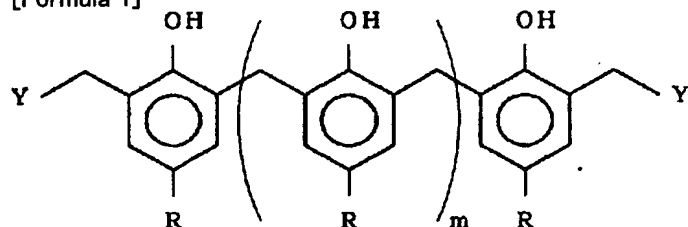
[0044] these organic peroxide — (\*\*) — olefin system copolymerization rubber and (\*\*) — the total quantity 100 weight section of a hydrogenation diene system polymer, an olefin (Ha) system polymer, a (d) ethylene system polymer, and an arbitration component like a postscript — receiving — the 0.02 - 1.5 weight section — the 0.05 - 1.0 weight section comes out comparatively preferably, and it blends, and is used. In under the 0.02 weight sections, if organic peroxide runs short of partial bridge formation of a copolymerization rubber component and exceeds the 1.5 weight sections on the other hand preferably, it becomes [ it becomes impossible to also hang down homogeneity and bridge formation / \*\*\*\* /, or decomposition to copolymerization rubber and a resinous principle, and ] a heterogeneous constituent and is not desirable.

[0045] moreover, the various above-mentioned bridge formation assistants — (\*\*) — olefin system copolymerization rubber and (\*\*) — it is preferably desirable for 0.2 - 2 weight section to come out comparatively, and to use to the total quantity 100 weight section of a hydrogenation diene system polymer, an olefin (Ha) system polymer, and an arbitration component like a postscript, below 3 weight sections. If there is more blending ratio of coal than this, when there are many loadings of organic peroxide, crosslinking reaction will progress, consequently equalization of a partial bridge formation mold thermoplastic-elastomer constituent will be lost, workability comes to fall, and, on the other hand, it remains in a partial bridge formation mold thermoplastic-elastomer constituent as a monomer unreacted when there are too few loadings of organic peroxide, and by the heat history at the time of carrying out fabrication of it, since change of physical properties is produced, superfluous use should be avoided. Moreover, two or more sorts of bridge formation assistants may be used together.

[0046] On the other hand, the phenol system resin cross linking agent used for this invention is matter expressed with the following general formula.

[0047]

[Formula 1]



[0048] Here, *m* is the integer of 0-10, *Y* is a hydroxyl group or a halogen atom, and *R* is the saturated hydrocarbon radical of

carbon numbers 1-15. Generally the above-mentioned matter is used as a rubber frame-common-equipment pons agent as indicated by for example, the U.S. Pat. No. 3,287,440 specification and this No. 3,709,840 specification. This cross linking agent is obtained by the condensation polymerization of a permutation phenol and an aldehyde in an alkali catalyst.

[0049] the amount of the above-mentioned phenol system resin cross linking agent used — (\*\*) — olefin system copolymerization rubber and (\*\*) — it is the range of 0.5 – 15 weight section to the total quantity 100 weight section of a hydrogenation diene system polymer, an olefin (Ha) system polymer, a (d) ethylene system polymer, and an arbitration component like a postscript. If oilproof [ of the constituent low / in dynamic bridge formation / and obtained ] and configuration recoverability are not enough and the amount of this phenol system resin cross linking agent used exceeds 15 weight sections on the other hand under in the 0.5 weight sections, the flexibility of the constituent obtained will be spoiled. The desirable amount of this cross linking agent used is the range of 2 – 8 weight section preferably to 1 – 10 weight section and a pan to the total quantity 100 weight section of each above-mentioned component. Although a phenol system resin cross linking agent can be used independently, in order to adjust a bridge formation rate, it can also be used together with a bridge formation accelerator. as a bridge formation accelerator — a stannous chloride, the metal halogenide of a ferric chloride, chlorination polypropylene, and bromination — organic halogenides, such as isobutylene isoprene rubber and chloroprene rubber, can be used. Moreover, it is more desirable if dispersants, such as a metallic oxide like a zinc oxide and stearin acid, are used.

[0050] In order to obtain the thermoplastic-elastomer constituent of this invention (\*\*) of a more than — a component and (\*\*) — besides a component, a component (Ha), a (d) component, and a (e) component The antioxidant of the amount of extent which does not check a mechanical strength, flexibility, and fabrication nature according to an application, An antistatic agent, a weathering agent, an ultraviolet ray absorbent, lubricant, an antiblocking agent, a seal nature amelioration agent, A crystalline-germ agent, a flameproofing agent, an antimicrobe and an antifungal agent, a tackifier, other softeners and plasticizers other than the above-mentioned straight-mineral-oil system softener, Coloring agents, such as titanium oxide and carbon black, a glass fiber, a carbon fiber, A metal fiber, an aramid fiber, a glass bead, a mica, a calcium carbonate, Gum polymers other than the above of bulking agents, such as a potassium titanate whisker, talc, a barium sulfate, a glass flake, and a fluororesin, an isobutylene-isoprene copolymer, etc., thermoplastics other than the above, etc. can be blended.

[0051] If good distribution of each component is obtained, manufacture of the thermoplastic-elastomer constituent of this invention may adopt what kind of approach, and will not be limited especially. Usually, melting kneading of each component is carried out with kneading machines, such as closed mold kneading machines, such as a roll mill used for rubber and resin industry, a Banbury mixer, and a pressurized kneader, or a 1 shaft extruder, and a 2 shaft extruder. Here, in order to heat-treat the above-mentioned constituent dynamically, it is usually desirable the temperature to which the component of (b) – (d) fuses the temperature (kneading temperature) of a kneading machine at least, and to carry out in 150–240 degrees C preferably 120–280 degrees C. Since early and distributing will become inadequate [ the reaction rate of a cross linking agent ] if kneading temperature becomes insufficient [ less than 120 degrees C / melting of a (b) – (d) component ] and it exceeds 280 degrees C on the other hand preferably, it is not desirable.

[0052] The thermoplastic-elastomer constituent of this invention can be used for components, such as packing of – exterior parts and weak-electric-current components, and housing, an industrial part, tarpaulin components, etc. among the automobiles by which conventional elasticity vinyl chloride system resin is used taking advantage of flexibility and fabrication nature being given, without spoiling a mechanical strength.

[0053]

[Example] Although an example is given and this invention is explained still more concretely hereafter, this invention is not restrained at all by the following examples, unless the summary is exceeded. In addition, among an example, the section and especially % are weight criteria, unless it refuses. Moreover, various kinds of measurement in an example was based on the following approaches.

[0054] After dissolving 5.0g of n-Deccan insoluble matter crystallinity alpha olefin system polymers in n-Deccan 150ml at the temperature of 150 degrees C and cooling to a room temperature, it cooled radiationally one whole day and night, and the ratio to the AUW of the alpha olefin system polymer in which centrifugal separation (for 23,000rpm and 30 minutes) was carried out twice, and was divided into, and the measured sludge (insoluble matter) weight dissolved the sludge (insoluble matter) was indicated by percent.

Number-average-molecular-weight number average molecular weight used trichlorobenzene as the solvent, and asked for it by polystyrene conversion using the gel permeation chromatography (GPC) in 135 degrees C.

[0055] Degree of hardness JIS (JIS A) It measured based on K6301.

Compression set JIS Based on K6301, it held under 25% compression for 22 hours, compression was canceled after that, and it asked from the recovery factor after 1-hour neglect. Elastic recovery nature is so good that a compression set is small.

100% modulus (M100), tensile strength, and maximum elongation JIS It measured based on K6301.

The fluidity was measured for the fluidity MFR (melt flow rate) on condition that the following.

temperature; — 230-degree-C load; — 10kg [0056] The polymers used in the example and the example of a comparison are as follows.

EP1 ethylene-propylene-ethylidene norbornene copolymerization rubber [72% of ethylene contents, a paraffin series oil 75phr oil exhibition, the Japan Synthetic Rubber Co., Ltd. make, and EP98A]

EP2 ethylene-propylene-dicyclopentadiene copolymerization rubber [70% of ethylene contents, the Japan Synthetic Rubber Co., Ltd. make, and EP75F]

EP3 ethylene-propylene-ethylidene norbornene copolymerization rubber [72% of ethylene contents, the Japan Synthetic Rubber Co., Ltd. make, and EP57P]

EP4 ethylene-propylene copolymerization rubber [73% of ethylene contents, the Japan Synthetic Rubber Co., Ltd. make, and EP07P]

EO1 ethylene and 1-octene copolymer [76% of ethylene contents, the Dow Chemical Co. make, and ENGAGE EG8100]



[0057] PP1 propylene-ethylene block polymer [93.5% of n-Deccan insoluble matter, the Mitsubishi Chemical make, and BC5C]  
 PP2 propylene-ethylene random polymer [92.2% of n-Deccan insoluble matter, the Mitsubishi Chemical make, and EX6]

PP3 polypropylene [98.7% of n-Deccan insoluble matter, the Mitsubishi Chemical make, and floor line6CK]

[0058] PE1 low density polyethylene [the Mitsubishi Chemical make and LJ800]

PE2 — a line — low density polyethylene [the Mitsubishi Chemical make and UF423]

PE3 high density polyethylene [the Mitsubishi Chemical make and FY50D]

PE4 ethylene and 1-hexene copolymer [the Mitsubishi Chemical make and SF240]

EO2 ethylene and 1-octene copolymer [the Dow Chemical Co. make and AFFINITYFM1570]

[0059] For the hydrogenation diene system polymer A hydrogenation diene system polymer A (henceforth "Polymer A"), it is a thing by Japan Synthetic Rubber Co., Ltd., and hydrogenation of the double bond of the butadiene part of the random copolymer of styrene and a butadiene is carried out, the vinyl joint content (1, 2-joint content) of the butadiene part in front of 4% and hydrogenation of a total combined styrene content is 80%, and total average molecular weight is the hydrogenation random copolymer of 300,000.

[0060] Hydrogenation diene system polymer B hydrogenation diene system polymer B (henceforth "Polymer B") it is a thing by Japan Synthetic Rubber Co., Ltd., and A-B-C structure [A is a polystyrene block, B is polybutadiene with many vinyl joint (1, 2-association) contents, and polybutadiene with little C to a vinyl joint content, and, as for B and C, hydrogenation of the double bond of the butadiene section is carried out — \*\*\*\* — having — the vinyl joint content of the B section — the vinyl joint content of 39% and the C section — 15% — it is — a total mean molecular weight — the hydrogenation block polymer of 150,000.

[0061] Hydrogenation diene system polymer C hydrogenation diene system polymer C (henceforth "Polymer C") it is a thing by Japan Synthetic Rubber Co., Ltd., and polybutadiene with little D-E-D-structure [D to a vinyl joint (1, 2-association) content and E are polybutadienes with many vinyl joint contents, and hydrogenation of the double bond of each butadiene part is carried out — \*\*\*\* — having — the vinyl joint content of the D section — the vinyl joint content of 15% and the E section — 35% — it is — a total mean molecular weight — the hydrogenation block polymer of 300,000.

[0062] Hydrogenation diene system polymer D hydrogenation diene system polymer D (henceforth "Polymer D") it is a thing by Japan Synthetic Rubber Co., Ltd., and polybutadiene with little D-E-D-structure [D to a vinyl joint (1, 2-association) content and E are polybutadienes with many vinyl joint contents, and hydrogenation of the double bond of each butadiene part is carried out — \*\*\*\* — having — the vinyl joint content of the D section — the vinyl joint content of 15% and the E section — 80% — it is — a total mean molecular weight — the hydrogenation block polymer of 300,000.

[0063] SEBS hydrogenation styrene-butadiene-styrene block polymer [the product made from Shell, and Krayton 1657]

SEPS hydrogenation styrene-isoprene-styrene block polymer [the Kuraray Co., Ltd. make and SEPTON2023]

[0064] Straight-mineral-oil system softener paraffin series softener [the Idemitsu Kosan make and PW-90]

Filler whiting [Maruo Calcium Co., Ltd. make, super S]

[0065] A cross linking agent 12, the 5-dimethyl -2, 5-di-tert-butyl peroxide-hexyne -3 [the Nippon Oil & Fats Co., Ltd. make and par hexyne 25B-40]

2 Ns of cross linking agents, N-m-phenylene bismaleimide [the product made from Ouchi Shinko Chemical Industry, and the BAL knock PM]

Cross linking agent 3 bromination alkylphenol formaldehyde [the product made from Taoka Chemistry, and the tacky roll 250]

[0066] Additive 1 zinc white [the product made from Sakai Chemistry, and a zinc white No. 1]

Additive 2 stearin acid [the Kao Corp. make and RUNAKKU S-30]

[0067] According to the following procedures, the constituent was prepared by the combination formula shown in one to examples 1-35 and example of comparison 15 tables 1-7. First, the temperature in a tub supplied the polymer, the softener, and the filler to the pressure type kneader [made in Moriyama Factory and 10l. of inner capacity] set as 180 degrees C. After carrying out preliminary mixing for 60 seconds, the bridge formation system was supplied, and it heat-treated dynamically for 480 seconds, and discharged. The discharged constituent was thrown into feeder RUDA [made in Moriyama Factory], the extruded strand was cut by the pelletizer, and the pellet was obtained.

[0068] The sheet of 2mm thickness was produced with the 0.5-oz injection molding machine, and the trial was presented with the obtained constituent (pellet). About evaluation of a degree of hardness (JIS A), a permanent set, and tensile strength, it pierced from the sheet to the predetermined test piece by the dumbbell cutter. Moreover, about evaluation of a compression set, using the test piece of 2mm thickness produced with the above-mentioned injection molding machine, after the punch, it adjusted so that it might become a regular dimension by pile, and the trial was presented. A result is shown in Tables 1-7.

[0069] The constituent of this invention is excellent in respect of the following so that clearly from the comparison with examples 1-36 and the examples 1-16 of a comparison. That is, its fluidity required for fabrication nature is also good while the thermoplastic-elastomer constituent of this invention discovers a flexible property and is excellent in elastic recovery nature, without spoiling a mechanical strength. On the other hand, since (b) and a (d) component are not contained, the constituent of the examples 1-9 of a comparison is inferior in the mechanical strength, when this degree of hardness compares. Moreover, since a modulus (M100) is high, flexibility is missing 100% and a hard feel is given, it is not desirable. The compounding ratio of this invention of a (b) component is [ the constituent of the example 10 of a comparison, and the example 11 of a comparison ] out of range, and its 100% modulus is high, and since the example 10 of a comparison gives a hard feel, the example 11 of a comparison is not [ that a fluidity is inferior and ] desirable [ the example ] preferably. Since the compounding ratio of this invention of a (b) component is out of range, the example 12 of a comparison does not have an inferior and desirable fluidity. The compounding ratio of this invention of a component (Ha) is out of range, and the example 13 of a comparison does not have an inferior and desirable fluidity. The compounding ratio of this invention of (b) and a (d) component is out of range, and since the example 14 of a comparison is inferior in flexibility, it is not desirable. The compounding ratio of this invention of a (e) component is out of range, and since the example 15 of a comparison is inferior in a mechanical strength and elastic recovery nature, it is

not desirable.

[0070]

[Table 1]

実施例	1	2	3	4	5	6	7	8	9
<b>配合処方 (部)</b>									
EP 1	53	53	53	53	60	40	30	60	46
重合体C	20	20	20	20	20	20	20	10	30
PP 1	9	9	13	18	5	10	20	10	8
PE 2	18	18	14	9	15	30	30	20	16
架橋剤 1	-	0.8	-	-	-	-	-	-	-
架橋剤 2	-	0.3	-	-	-	-	-	-	-
架橋剤 3	1.0	-	1.0	1.0	1.0	1.0	1.0	1.0	1.0
添加剤 1	1.0	-	1.0	1.0	1.0	1.0	1.0	1.0	1.0
添加剤 2	0.3	-	0.3	0.3	0.3	0.3	0.3	0.3	0.3
<b>物性</b>									
硬度 (JIS A)	75	78	78	80	69	85	90	78	75
圧縮永久歪 (%)	30	29	35	36	27	35	37	40	28
M100(kgf/cm <sup>2</sup> )	32	35	36	38	26	45	56	35	31
引張強さ (kgf/cm <sup>2</sup> )	145	135	125	120	125	165	180	125	156
最大伸び (%)	760	610	730	700	780	700	680	700	780
MFR (g/10min)	6.6	4.3	10.3	13.5	4.7	8.4	10.3	3.8	8.3

[0071]

[Table 2]

実施例	10	11	12	13	14
<b>配合処方 (部)</b>					
EP 1	30	30	30	30	20
重合体 C	20	20	10	10	40
PP 1	10	40	10	50	10
PE 2	40	10	50	10	20
架橋剤 1	-	-	-	-	-
架橋剤 2	-	-	-	-	-
架橋剤 3	1.0	1.0	1.0	1.0	1.0
添加剤 1	1.0	1.0	1.0	1.0	1.0
添加剤 2	0.3	0.3	0.3	0.3	0.3
<b>物性</b>					
硬度 (J I S A)	87	95	92	95	90
圧縮永久歪 (%)	34	41	38	38	39
M100 (kgf/cm <sup>2</sup> )	46	70	48	50	43
引張強さ (kgf/cm <sup>2</sup> )	210	140	180	130	250
最大伸び (%)	710	450	650	420	610
MFR (g/10min)	8.7	15.3	9.3	18.9	9.8

[0072]

[Table 3]

実施例	15	16	17	18	19	20	21	22	23	24
<b>配合処方 (部)</b>										
EP 1	53	53	53	53	53	53	-	-	-	-
EP 2	-	-	-	-	-	-	53	-	-	-
EP 3	-	-	-	-	-	-	-	53	-	-
EP 4	-	-	-	-	-	-	-	-	53	-
EO 1	-	-	-	-	-	-	-	-	-	53
重合体 C	20	20	20	20	20	20	20	20	20	20
PP 1	9	9	9	9	-	-	9	9	9	9
PP 2	-	-	-	-	9	-	-	-	-	-
PP 3	-	-	-	-	-	9	-	-	-	-
PE 1	18	-	-	-	-	-	-	-	-	-
PE 2	-	-	-	-	18	18	18	18	18	18
PE 3	-	18	-	-	-	-	-	-	-	-
PE 4	-	-	18	-	-	-	-	-	-	-
EO 2	-	-	-	18	-	-	-	-	-	-
架橋剤 1	-	-	-	-	-	-	-	-	0.8	0.8
架橋剤 2	-	-	-	-	-	-	-	-	0.3	0.3
架橋剤 3	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	-	-
添加剤 1	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	-	-
添加剤 2	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	-	-
<b>物性</b>										
硬度 (JIS A)	74	77	75	74	75	77	83	84	82	88
圧縮永久歪 (%)	28	31	31	29	32	35	44	45	42	48
M100 (kgf/cm <sup>2</sup> )	31	33	32	33	35	27	20	22	21	22
引張強さ (kgf/cm <sup>2</sup> )	155	140	120	150	150	130	135	140	110	125
最大伸び (%)	750	730	740	780	750	740	620	650	890	910
MFR (g/10min)	5.3	6.2	8.6	7.1	5.9	5.6	2.3	2.1	1.8	2.0

[0073]

[Table 4].

実施例	25	26	27	28	29	30	31	32
<b>配合処方 (部)</b>								
EP1	53	53	53	53	53	53	53	53
重合体A	-	-	20	-	-	-	-	-
重合体B	-	-	-	20	-	-	-	-
重合体C	-	10	-	-	-	-	20	20
重合体D	20	10	-	-	-	-	-	-
SEBS	-	-	-	-	20	-	-	-
SEPS	-	-	-	-	-	20	-	-
PP1	9	9	9	9	9	9	9	9
PE2	18	18	18	18	18	18	18	18
架橋剤3	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
添加剤1	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
添加剤2	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3
鉱物油系軟化剤	-	-	-	-	-	-	20	50
フィラー	-	-	-	-	-	-	-	-
<b>物性</b>								
硬度 (JIS A)	71	73	68	75	75	75	69	58
圧縮永久歪 (%)	31	30	40	34	36	38	25	33
M100 (kgf/cm <sup>2</sup> )	30	31	27	35	35	35	29	20
引張強さ (kgf/cm <sup>2</sup> )	110	135	145	132	130	125	120	98
最大伸び (%)	820	800	760	780	740	780	910	500
MFR (g/10min)	8.2	7.3	6.6	5.3	5.8	5.4	8.2	15.3

[0074]  
[Table 5]

実施例	33	34	35
<u>配合処方 (部)</u>			
EP1	53	53	53
重合体A	-	-	-
重合体B	-	-	-
重合体C	20	20	20
重合体D	-	-	-
SEBS	-	-	-
SEPS	-	-	-
PP1	9	9	9
PE2	18	18	18
架橋剤3	1.0	1.0	1.0
添加剤1	1.0	1.0	1.0
添加剤2	0.3	0.3	0.3
鉱物油系軟化剤	-	20	100
フィラー	30	30	-
<u>物性</u>			
硬度 (JIS A)	77	71	42
圧縮永久歪 (%)	32	28	31
M100 (kgf/cm <sup>2</sup> )	37	31	15
引張強さ (kgf/cm <sup>2</sup> )	125	125	86
最大伸び (%)	700	870	800
MFR (g/10min)	6.1	7.6	45.3

[0075]  
[Table 6]

比較例	1	2	3	4	5	6	7	8	9
<b>配合処方 (部)</b>									
EP 1	60	70	70	80	-	-	-	70	70
EP 2	-	-	-	-	70	-	-	-	-
EP 3	-	-	-	-	-	70	-	-	-
EP 4	-	-	-	-	-	-	70	-	-
PP 1	40	30	30	20	30	30	30	-	-
PP 2	-	-	-	-	-	-	-	30	-
PP 3	-	-	-	-	-	-	-	-	30
架橋剤 1	-	-	0.8	-	-	-	0.8	-	-
架橋剤 2	-	-	0.3	-	-	-	0.3	-	-
架橋剤 3	1.0	1.0	-	1.0	1.0	1.0	-	1.0	1.0
添加剤 1	1.0	1.0	-	1.0	1.0	1.0	-	1.0	1.0
添加剤 2	0.3	0.3	-	0.3	0.3	0.3	-	0.3	0.3
<b>物性</b>									
硬度 (JIS A)	94	85	83	76	88	88	86	84	86
圧縮永久歪 (%)	32	28	36	27	33	34	38	30	31
M100(kgf/cm <sup>2</sup> )	80	65	60	51	66	63	62	61	62
引張強さ (kgf/cm <sup>2</sup> )	100	85	75	68	70	75	65	80	85
最大伸び (%)	420	430	480	510	410	430	490	450	400
MFR (g/10min)	4.3	3.6	9.2	2.3	2.1	2.3	7.6	3.1	2.3

[0076]  
[Table 7]

比較例	10	11	12	13	14	15
<b>配合処方 (部)</b>						
EP 1	5	94	5	60	1	53
重合体 C	40	3	90	20	2	20
PP 1	30	2	2	-	3	9
PE 2	25	1	3	20	95	18
架橋剤 3	1.0	1.0	1.0	1.0	1.0	1.0
添加剤 1	1.0	1.0	1.0	1.0	1.0	1.0
添加剤 2	0.3	0.3	0.3	0.3	0.3	0.3
鉱物油系軟化剤	-	-	-	-	-	250
<b>物性</b>						
硬度 (JIS A)	96	35	89	65	98	30
圧縮永久歪 (%)	38	24	25	30	80	68
M100 (kgf/cm <sup>2</sup> )	120	20	90	35	78	10
引張強さ (kgf/cm <sup>2</sup> )	130	115	185	98	120	35
最大伸び (%)	350	610	510	420	390	1200
MFR (g/10min)	2.1	0.01	0.01	0.1	9.3	800

[0077]

[Effect of the Invention] Its balance of a mechanical strength and elastic recovery nature is also good while the thermoplastic-elastomer constituent of this invention is excellent in flexibility and a fluidity. Moreover, the modulus in the small part of elongation is small, and it is possible to give a flexible feel. Therefore, it can be used for components, such as packing housing of - exterior parts and weak-electric-current components, an industrial part, tarpaulin components, gasket seal components, etc. among the automobiles by which elasticity vinyl chloride system resin is used.

[Translation done.]



\* NOTICES \*

Japan Patent Office is not responsible for any damages caused by the use of this translation.

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. \*\*\*\* shows the word which can not be translated.
3. In the drawings, any words are not translated.

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CORRECTION OR AMENDMENT

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[Kind of official gazette] Printing of amendment by the convention of 2 of Article 17 of Patent Law  
[Section partition] The 3rd partition of the 3rd section  
[Publication date] July 19, Heisei 14 (2002. 7.19)

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C08L 23/00 LCD  
LCB  
C08K 5/01 KEH  
5/14 KES  
C08L 15/00 LBG  
23/04 LDD  
// C08F 8/00 MHZ

[FI]

C08L 23/00 LCD  
LCB  
C08K 5/01 KEH  
5/14 KES  
C08L 15/00 LBG  
23/04 LDD  
C08F 8/00 MHZ

[Procedure revision]  
[Filing Date] April 26, Heisei 14 (2002. 4.26)  
[Procedure amendment 1]  
[Document to be Amended] Specification  
[Item(s) to be Amended] Claim  
[Method of Amendment] Modification  
[Proposed Amendment]  
[Claim(s)]  
[Claim 1] (b) 10 - 90 % of the weight of olefin system copolymerization rubber,  
(b) 1 - 80 % of the weight of hydrogenation diene system polymers,  
(c) 1 - 80 % of the weight of crystalline alpha olefin system polymers which use a with a carbon numbers of three or more alpha olefin as a principal component,  
(d) 1 - 80 % of the weight of ethylene system polymers  
[ -- however, (\*\*) -- + (\*\*) -- +(Ha)+ (d) = 100-% of the weight] -- and  
(e) About a straight-mineral-oil system softener, it is the 0 - 200 weight section to the total quantity 100 weight section of an above-mentioned (b) - (d) component.  
The thermoplastic-elastomer constituent which comes to heat-treat the blended constituent under existence of a rubber frame-common-equipment pons agent dynamically.  
[Claim 2] (b) The thermoplastic-elastomer constituent according to claim 1 characterized by a component being the hydrogenation diene system polymer which is the following (low 1), or (low 3) is shown.  
(\*\*1) A component is a hydrogenation diene system polymer obtained by hydrogenating the polymer which makes a subject the random copolymerization parts of conjugated diene and an aromatic series vinyl compound.  
(\*\*3) The polybutadiene polymer block whose (D) vinyl joint content of a hydrogenation diene system polymer is 25% or less, (E) They are a conjugated diene polymer block or a copolymer block with an aromatic series vinyl compound and conjugated diene. the polymer block whose vinyl joint content of a conjugated diene part is 25 - 95% -- (D)-(E)-(D) or (D)- it is obtained by hydrogenating the double bond part of the block copolymer of the shape of a straight chain arranged as shown in (E), and the

letter of branching 80% or more.

[Claim 3] (b) The thermoplastic-elastomer constituent according to claim 1 or 2 which is the denaturation hydrogenation diene system polymer with which the component denaturalized by the functional group.

[Claim 4] The thermoplastic-elastomer constituent according to claim 1 to 3 whose rubber frame-common-equipment pons agent is organic peroxide or a phenol system resin cross linking agent.

[Procedure amendment 2]

[Document to be Amended] Specification

[Item(s) to be Amended] 0007

[Method of Amendment] Modification

[Proposed Amendment]

[0007] In these (b) olefin system copolymerization rubber, it is desirable that copolymerization of the with a carbon numbers of three or more alpha olefin is carried out at 10% of the weight or more of a rate. Moreover, as nonconjugated diene, ethylidene norbornene, dicyclopentadiene, 1, and 4-hexadiene is desirable, it is the amount which becomes 40 or less by iodine number display, and existing is desirable.

(b) In olefin system copolymerization rubber, the content of an ethylene component increases more than 90 % of the weight, and the content of an alpha olefin component runs short [ the flexibility of this copolymerization rubber ] and is not desirable at less than 10 % of the weight.

moreover, (\*\*) -- as for the Mooney viscosity (1+4,100 degree C of ML(s)) of olefin system copolymerization rubber, it is desirable 10-500, and that it is 30-400 preferably. If this Mooney viscosity is smaller than 10, reinforcement will become low, and on the other hand, if larger than 500, a maldistribution with (\*\*) and (Ha) a (d) component generates and is not desirable.

[Procedure amendment 3]

[Document to be Amended] Specification

[Item(s) to be Amended] 0017

[Method of Amendment] Modification

[Proposed Amendment]

[0017] In the hydrogenation (low 1) diene system polymer of this invention, the rate of hydrogenation of the double bond of the conjugated diene part in a chain is 90% or more especially preferably 85% or more still more preferably 80% or more preferably. At less than 80%, weatherability becomes inadequate. Moreover, number average molecular weight will be 50,000-600,000 preferably [ it is desirable and ] to 50,000-700,000, and a pan, and less than by 50,000, the hydrogenation (low 1) diene system polymer of this invention has an inadequate mechanical strength, on the other hand, if 700,000 is exceeded, it will run short of a fluidity and will become what has inadequate fabrication nature.

(\*\*~1) A component can be obtained by the approach currently indicated by JP,3-72512,A.

[Procedure amendment 4]

[Document to be Amended] Specification

[Item(s) to be Amended] 0020

[Method of Amendment] Modification

[Proposed Amendment]

[0020] Moreover, the (B) block which constitutes a hydrogenation (low 2) diene system polymer is a conjugated diene block (\*\*) polymer, and 30 to 90% of the weight, it is 35 - 80 % of the weight still more preferably, and at less than 30 % of the weight, if flexibility, on the other hand, exceeds 90 % of the weight by falling, as for the desirable content of the (B) block, workability and a mechanical strength will fall. (B) The vinyl joint content of the conjugated diene part before the hydrogenation included in a block is 30 - 90% still more preferably 25 to 95% preferably. (B) If hydrogenation of the vinyl joint content is carried out at less than 25% when conjugated diene is a butadiene among the conjugated diene blocks before the hydrogenation which is blocked, a polyethylene chain will generate, a rubber-property will be lost, if it exceeds 95% on the other hand and hydrogenation will be carried out, glass transition temperature will become high, a rubber-property is lost, and it is not desirable. (B) the desirable number average molecular weight of a block -- desirable -- 15,000- 630,000 -- further -- desirable -- 35,000- it is 420,000. Moreover, the rate of hydrogenation of the double bond of the conjugated diene part of the (B) block is 80% or more.

[Procedure amendment 5]

[Document to be Amended] Specification

[Item(s) to be Amended] 0025

[Method of Amendment] Modification

[Proposed Amendment]

[0025] The aromatic series vinyl compound used here in order to obtain a component (low 3), and conjugated diene are the same as that of what is used for a component (low 1).

(\*\*~3) The (D) block in a component turns into a crystalline polymer block which shows structure similar to usual low density polyethylene (LDPE) by hydrogenation. (D) Although the vinyl joint content under block is 25% or less, it is usually 15% or less still more preferably 20% or less preferably. (D) When the vinyl joint content under block exceeds 25%, descent of the crystalline melting point after hydrogenation is remarkable, and a mechanical property is inferior.

[Procedure amendment 6]

[Document to be Amended] Specification

[Item(s) to be Amended] 0037

[Method of Amendment] Modification

[Proposed Amendment]

[0037] Here, the rubber elasticity and flexibility of a constituent which are acquired as the loadings of (b) olefin system copolymerization rubber are less than 10 % of the weight come to get worse. On the other hand, if a (b) component exceeds 90 %

of the weight, the fabrication nature of the constituent obtained and a mechanical strength will get worse. Moreover, the elastic recovery nature and the mechanical strength of a constituent from which a (b) component is obtained at less than 1 % of the weight are inferior, and on the other hand, when it exceeds 80 % of the weight, the thermal resistance and the fluidity of a constituent which are acquired are inferior. Furthermore, the thermal resistance of the constituent obtained as a component (Ha) is less than 1 % of the weight gets worse, and on the other hand, if it exceeds 80 % of the weight, the flexibility of the constituent obtained will get worse. Furthermore, the elastic recovery nature and the mechanical strength of a constituent from which a (d) component is obtained at less than 1 % of the weight are inferior, and on the other hand, if it exceeds 80 % of the weight, the flexibility of the constituent obtained runs short.

[Procedure amendment 7]

[Document to be Amended] Specification

[Item(s) to be Amended] 0043

[Method of Amendment] Modification

[Proposed Amendment]

[0043] If a suitable bridge formation assistant is made to exist with organic peroxide, homogeneity and crosslinking reaction [ \*\*\*\* ] are expectable. As a bridge formation assistant, sulfur, p-quinonedioxime, p, and p'-dibenzoyl quinonedioxime, Ethylene glycol dimethacrylate, diethylene-glycol dimethacrylate, Triethylene glycol dimethacrylate, tetra-ethylene dimethacrylate, Polyethylene glycol dimethacrylate, trimethylolpropanetrimethacrylate, Diaryl phthalate, tetra-aryloxy ethane, thoria reel SHIANU rate, N, and N-m-phenylene bismaleimide, a maleic anhydride, a divinylbenzene, JI (meta) acrylic-acid zinc, etc. are used. Preferably, it is desirable to use N and N-m-phenylene bismaleimide, p, and p'-dibenzoyl quinonedioxime and a divinylbenzene. Moreover, it can also use as a cross linking agent by the N and N-m-phenylene bismaleimide independent.

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[Translation done.]